



## Use of Personal Measurements for Ozone Exposure Assessment: A Pilot Study

L.-J. Sally Liu,<sup>1</sup> Petros Koutrakis,<sup>1</sup> Helen H. Suh,<sup>1</sup> James D. Mulik,<sup>2</sup> and Robert M. Burton<sup>2</sup>

<sup>1</sup>Department of Environmental Health, Harvard School of Public Health, Boston, MA 02115 USA; <sup>2</sup>Atmospheric Research and Exposure Assessment Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711 USA

During summer 1991, we collected indoor, outdoor, and personal ozone concentration data as well as time-activity data in State College, Pennsylvania. These concentrations were measured for 23 children and their homes using passive ozone samplers. Outdoor concentrations were also measured at a stationary ambient monitoring site. Results from this pilot study demonstrate that fixed-site ambient measurements may not adequately represent individual exposures. Outdoor ozone concentrations showed substantial spatial variation between rural and residential regions. Ignoring this spatial variation by using fixed-site measurements to estimate personal exposures can result in an error as high as 127%. In addition, evidence from our pilot study indicates that ozone concentrations of a single indoor microenvironment may not represent those of other indoor microenvironments. Personal exposures were significantly correlated with both indoor ( $r = 0.55$ ) and outdoor ( $r = 0.41$ ) concentrations measured at home sites. Multiple regression analyses identified indoor ozone concentrations as the most important predictors of personal exposures. However, models based on time-weighted indoor and outdoor concentrations explained only 40% of the variability in personal exposures. When the model included observations for only those participants who spent the majority of their day in or near their homes, an  $R^2$  of 0.76 resulted when estimates were regressed on measured personal exposures. It is evident that contributions from diverse indoor and outdoor microenvironments must be considered to estimate personal ozone exposures accurately. **Key words:** exposure modeling, ozone, passive sampler, personal exposure assessment. *Environ Health Perspect* 101:318–324(1993)

Acute and chronic lung function responses to ozone exposure have been investigated extensively in a variety of epidemiological studies (1–14). These studies, however, have a limited role in determining accurate dose–response relationships for ozone (15,16). In most epidemiological studies, population ozone exposures are assumed to be identical to the concentrations measured at an ambient monitoring site. Lebowitz et al. (2) found that this

assumption may be flawed and concluded that personal ozone exposures may be very different from those measured at both outdoor and indoor monitoring sites. Fixed-location measurements do not account for the effects of spatial variation in ozone concentrations, indoor/outdoor concentration differences (17–22), and varying activity patterns on personal exposures (21).

A major limitation of previous ozone exposure investigations is the lack of a personal exposure or microenvironmental ozone monitor. With the recent development of an ozone passive sampler by Koutrakis et al. (23), personal, indoor microenvironmental, and outdoor concentrations can be measured on a wide scale. This sampler has been validated in a variety of laboratory conditions for temperature, relative humidity, wind velocity, ultraviolet radiation, and other atmospheric oxidant interferences. Because of its low cost and small size (weight = 7 g, size = 2 cm diameter  $\times$  3 cm), the passive sampler is especially suited for characterizing the exposure pattern of individuals in large-scale epidemiological studies.

This paper describes a pilot study conducted during summer 1991, in State College, Pennsylvania, to assess ozone exposures using the passive ozone sampler. This pilot study, which was performed in conjunction with an acid aerosol monitoring study, enhances our understanding of ozone concentrations in various outdoor and indoor environments and characterizes individual ozone exposures. During the study, extensive personal measurements and detailed time-activity information were collected for 23 children, and indoor and outdoor concentrations were measured at their homes. Additional outdoor measurements were taken at a stationary monitoring site. Factors affecting variation of indoor and outdoor ozone concentrations as well as personal ozone exposures were examined. Multivariate regression models and simple microenvironmental exposure models were developed to provide a practical means for estimating personal exposures.

## Methods

Ozone concentrations were measured in State College, Pennsylvania, a college town located approximately 240 km east of Pittsburgh, with a population of 36,000. Indoor, outdoor, and personal monitoring was performed for 23 children (ages 10–11). All of these children lived in non-smoking households in one of six residential regions. Except for two children who lived in apartment buildings, all the children lived in single-family residences. Homes were located at altitudes ranging from 200 to 400 m. Two of the participants' homes had gas stoves and 12 had air conditioners. However, only three of these homes used air conditioning during the monitoring period. Of all the sampled homes, 13 (including one air-conditioned home) used fans in the sampling room for cooling, and the others used either air conditioning (2 homes) or open windows (8 homes).

We also measured outdoor ozone concentrations at a stationary ambient monitoring (SAM) site, located at the State College National Dry Deposition Network Scotia Range site, approximately 6 km west of downtown State College. Measurements from the SAM site served as reference levels for comparisons to home-site concentrations.

We monitored ozone from 8 July–27 August 1991. In general, three children and their homes were monitored each week for up to 6 days. For each home, a maximum of six 12-hr indoor daytime, five 12-hr indoor nighttime, four 24-hr out-

Address correspondence to L.-J.S. Liu, Department of Environmental Health, Harvard School of Public Health, Building I, G-13B, 665 Huntington Avenue, Boston, MA 02115 USA. Part of this study has been funded by the U.S. Environmental Protection Agency under cooperative agreement to Harvard School of Public Health no. CR816 740-02. This paper has been subjected to EPA peer and administrative review, and it has been approved for publication as an EPA document. Mention of trade names or commercial products does not constitute endorsement or recommendation for use. This work was also supported by the Electric Power Research Institute (contract no. RP 1630-59). We acknowledge Mary Ann Allan and Janice Yager of EPRI for their contributions. This study could not have been done without the intensive field work of Mark Davey, Dave Decapria, Carolyn Shively, Jeffrey Myers, Erik Selekman, Glenn Hunter, and Robert Surh. Ben Rosenthal's dedicated lab analysis is highly appreciated. Discussions with Mike Wolfson, David Wypij, Louise Ryan, P. Barry Ryan, George Allen, and Sue Froehlich of the Harvard School of Public Health were extremely helpful.

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door, one 12-hr daytime outdoor, and six 12-hr personal daytime samples were collected (Fig. 1). For logistical reasons, we collected the 12-hr home outdoor samples at the end of each monitoring week. In addition, 12-hr daytime and nighttime outdoor passive samples were taken at the SAM site each day. Indoor, outdoor, and personal samples were collected simultaneously, with sampling times beginning or ending at 8 AM or 8 PM.

For indoor sampling, passive samplers were clipped on a camera tripod and placed in the main activity rooms of children's homes, at least 1 m from walls, windows, air conditioners, and other ventilation devices to avoid excess air flow. Samplers were located 1.2 m above the floor, so that ozone concentrations were measured at about the height of a child's breathing zone.

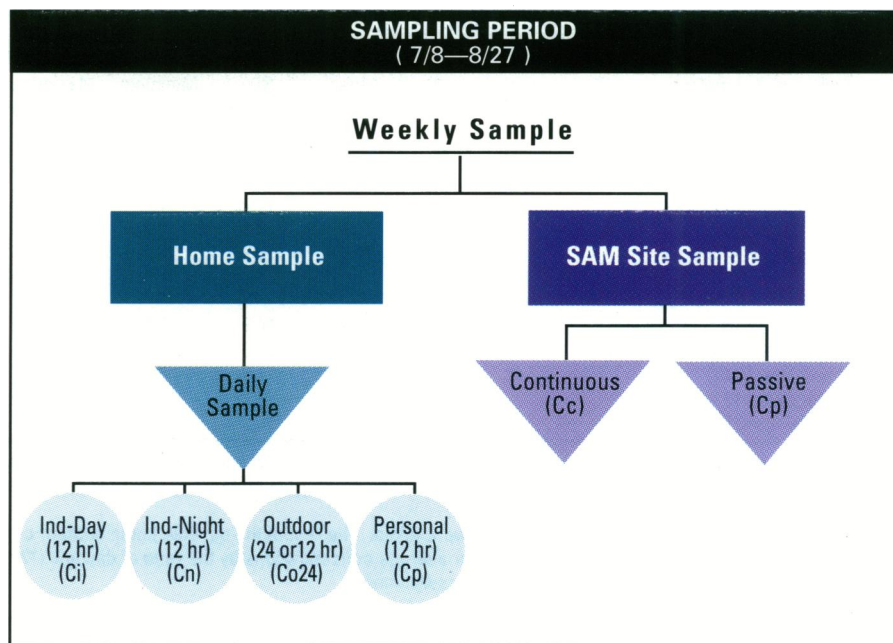
A questionnaire regarding the ventilation conditions of the homes, including use of air conditioning, hours of windows/doors opened, and percentage and location of the open windows/doors, was administered at the end of each sampling day. We also measured air exchange rates in each home using the perfluorocarbon tracer gas method (24). Results from this analysis will be discussed in forthcoming papers (Liu et al., in preparation).

For outdoor sampling, on each sampling day, we suspended one passive sampler under a protective cup, which was attached to a tripod in the front or backyard of each home. The protective cups were made of opaque white polyvinyl chloride (PVC) pipe. The cups were used to minimize face-velocity effects on the sampler collection rate and to protect the samplers from rain (23). We placed samplers approximately 1.2 m above ground to measure ozone concentrations at about the level of a child's breathing zone.

The same sampling procedure was performed at the SAM site: one passive sampler was placed under a PVC protective cup. The passive sampler was co-located with the manifold inlet of a UV photometric ozone analyzer at a height of 3.5 m.

For personal sampling, we clipped personal samplers onto the strap of a backpack worn by each participant. (The backpack was used to hold a pump and a timer for the concurrent acid aerosol measurements.) Samplers were placed at chest level to correspond to breathing height. Participants were asked to wear the backpack throughout the 12-hr daytime monitoring period.

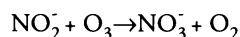
On each day, participants recorded their activities and the amount of time spent in different environments. To assure maximum accuracy and compliance, we aggregated these records into 30-min periods and field technicians transferred them



**Figure 1.** Monitoring plan and sampling duration. Typically, three children and their homes were sampled each week, in addition to the stationary ambient monitoring (SAM) site samples. Ind, indoor.

onto formatted time-activity sheets with the help of the parents and children at the end of each day. On the formatted time-activity sheet, locations were divided into four groups: home, near home (within a few blocks of home), school, and other.

The passive ozone sampler, developed by Koutrakis et al. (23), consists of a badge clip supporting a barrel-shaped device developed by Ogawa and Company (Pompano Beach, Florida). The sampler contains two glass-fiber filters coated with potassium carbonate ( $K_2CO_3$ ) and sodium nitrite ( $NaNO_2$ ). The sampling technique is based on the oxidation reaction of nitrite ( $NO_2^-$ ) by ozone to produce nitrate ( $NO_3^-$ ):



The amount of nitrate is determined using ion chromatography. The average ozone concentration is calculated from the measured nitrate concentration and a previously determined collection rate (25.5 cc/min) (23). The passive samplers performed well in controlled laboratory tests at typical ambient ozone levels (40 ppb–100 ppb) under relative humidities and temperatures varying from 10 to 80% and 0 to 40°C, respectively (23). The limit of detection (LOD), defined as three times the standard deviation of the field blanks used in this study, was 18 ppb for 12-hr measurements.

A UV photometric ozone analyzer (Thermo-Electron Co. Model 49) used at the SAM site is designated as an equivalent method for ambient ozone measurements by the U.S. Environmental Protection Agency. The LOD of the UV method is 2

ppb, and the precision is 2 ppb (25). We calibrated the continuous analyzer once a week with span checks with 0, 50, and 400 ppb ozone, and once a month with 0, 50, 100, 200, and 400 ppb of ozone.

We used SAS (26) software for all data management and statistical analyses. Data capture rate was 85% of all possible samples. We analyzed results in five ways. 1) Simple regression analysis and calculation of Pearson's correlation coefficient for passive and continuous ozone measurements were performed. The relative error of passive sampler measurements to continuous measurements was also determined. (For all analyses except for the evaluation of the passive sampler, we used continuous measurements as the SAM site ozone concentrations.) 2) Two-sample *t*-tests and one-way analysis of variance (ANOVA) techniques were used to determine whether outdoor concentrations varied spatially. 3) Diurnal variation in outdoor and indoor ozone concentrations was determined using two-sample *t*-tests, comparing the means of daytime and nighttime concentrations. 4) The ratio of indoor to outdoor ozone concentration (I/O ratio) was calculated for each home. The variation in I/O ratios among homes was further examined using ANOVA techniques. 5) Personal exposures were compared to indoor and outdoor measurements using paired *t*-tests and correlation analyses. We developed personal exposure models using multiple linear regression analyses and the time-weighted microenvironmental concept. The *p*-values associated with the tests of significance of regression coefficients serve only as a guide because autocorrelation



**Table 1.** Summary of ozone concentration measurements at State College, Pennsylvania, summer 1991

Location/type of samples	N	Mean	SD	Minimum	Maximum
<b>Stationary ambient monitoring site</b>					
Passive daytime (12 hr) <sup>a</sup>	47	56.4	16.2	30.6	94.5
Passive nighttime (12 hr)	50	19.1	8.9	3.3	40.1
Continuous daytime (12 hr)	47	55.3	14.7	27.8	92.3
Continuous nighttime (12 hr)	50	20.1	10.1	3.2	44.1
Continuous daily (24 hr)	46	37.8	10.7	18.3	64.3
<b>Home site</b>					
Outdoor daily (24 hr)	68	29.8	14.3	7.0	64.1
Estimated outdoor daytime (12 hr) <sup>b</sup>	77	45.9	21.3	11.6	104.1
Indoor daytime (12 hr)	84	19.2	11.3	0.8	46.0
Indoor nighttime (12 hr)	65	10.5	7.2	0.5	32.9
Personal daytime (12 hr)	81	23.9	16.2	0.5	78.8

<sup>a</sup>Four daytime samples were detected as outliers using the simple residual method at a 99% confidence level. These four outliers are presumably due to analytical laboratory mistakes and have been removed from the data set.

<sup>b</sup>Nine 12-hr (daytime) home site outdoor samples were taken. The rest of the daytime home outdoor concentrations were estimated by multiplying the 24-hr average concentrations by the ratio of daytime (12-hr average) to 24-hr average SAM site continuous measurements.

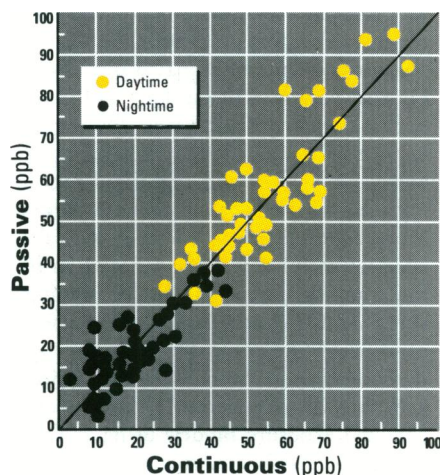
**Table 2.** Relative error of passive sampler measurements at the stationary ambient monitoring site

Range of concentration (ppb)	N	Relative error <sup>a</sup> (%)	Uncertainty <sup>b</sup> (ppb)
0 ≤ O <sub>3</sub> < 10	11	90	4.5
10 ≤ O <sub>3</sub> < 20	18	30	4.5
20 ≤ O <sub>3</sub> < 30	12	24	6.0
30 ≤ O <sub>3</sub> < 40	12	16	5.6
40 ≤ O <sub>3</sub> < 50	17	16	7.2
50 ≤ O <sub>3</sub> < 60	11	12	6.6
60 ≤ O <sub>3</sub> < 70	10	18	11.7
70 ≤ O <sub>3</sub> < 80	3	9	6.8
80 ≤ O <sub>3</sub> < 95	3	10	8.7

<sup>a</sup>The relative error is defined as the root mean square difference of the passive ( $C_{ps,i}$ ) and continuous ( $C_{c,i}$ ) measurements divided by the mean of the continuous measurements ( $C_{cc}$ ):

$$\sqrt{\frac{\sum_{i=1}^N (C_{c,i} - C_{ps,i})^2}{N}} \div C_{cc}$$

<sup>b</sup>Uncertainty = relative error mean × range concentration (ppb).

**Figure 2.** Ozone concentrations measured by passive samplers and the continuous analyzer at the stationary ambient monitoring site. Note that the graph is overlaid by a 45° line.

± 22% of their time inside their homes, 11 ± 12% of the time inside other microenvironments, and 30 ± 22% of the time outdoors.

The ozone concentrations measured with the passive samplers at the SAM site were in excellent agreement with those measured by the co-located continuous monitor (Fig. 2). The Pearson's correlation coefficient for the passive and continuous measurements was 0.95 ( $p < 0.01$ ). The relative error of the passive measurements to the continuous measurements at the SAM site decreased with increasing ozone concentrations (Table 2). For measurements below or near the LOD, the relative errors reflect an uncertainty of only 4.5 ppb (i.e.,  $0.90 \times 5$  or  $0.30 \times 15$  ppb). In general, the uncertainty of the passive sampler measurements was well below 10 ppb.

### Outdoor Spatial and Diurnal Variation

Outdoor (24 hr) ozone concentrations measured at home sites were highly correlated with the SAM site ozone concentrations ( $r = 0.81$ ,  $p < 0.01$ ). Despite this agreement, there was a substantial difference in ozone concentrations between the SAM site and home outdoor sites. The mean outdoor concentration at the SAM site ( $37.8 \pm 10.7$  ppb) was significantly higher than that for home sites ( $29.8 \pm 14.3$ ) using a two-sample  $t$ -test ( $p < 0.01$ ). The mean ratio of home to SAM site outdoor (24 hr) concentrations was  $0.80 \pm 0.25$ .

Spatial variation in outdoor concentrations was also observed when homes were grouped into six residential regions. Region 1, which includes downtown State College, has the greatest home, population, and traffic density. Regions 2–5 are populated residential areas but less dense compared to region 1. Region 6 is the least densely populated community. The mean ratio of outdoor home to SAM site concentration varied significantly by region (Table 3) using ANOVA techniques ( $F$ -value = 3.06,  $p < 0.05$ ). When the mean ratios were further examined using Tukey's pairwise comparison method at the 95% confidence level, the mean ratio of the most rural area, region 6, was significantly higher than those for the densely populated regions 1 and 4.

Altitude of the home sites (Table 3) was not correlated with the observed spatial variation ( $r = 0.04$ ,  $p = 0.75$ ). Spatial variation more likely resulted from differences in home density and traffic. Higher density of homes may provide greater surface area for ozone depletion, whereas higher traffic density may increase NO concentrations, which reacts with ozone

between repeated measurements on the same subjects over time was not considered.

### Results

A summary of results for samples collected is presented in Table 1. We collected 47 daytime and 50 nighttime outdoor passive samples at the SAM site. At home sites, we collected 68 outdoor (24 hr), 84 indoor daytime, 65 indoor nighttime, and 81 personal daytime samples. Simultaneous continuous measurements were also tabulated. For comparison purposes, 24-hr continuous measurements at the SAM site and estimated 12-hr home outdoor daytime concentrations are also listed in Table 1.

In addition to ozone measurements, we collected 94 time-activity diaries from the 23 participants during daytime sampling periods. On average, participants spent 59

(27). This theory is supported by the similarity of the ozone concentrations between region 6 and the SAM site. Even though these sites are 13 km apart, their concentrations are comparable, with a mean ratio approximately equal to 1.

Both indoor and outdoor ozone concentrations exhibited a diurnal pattern (Table 1), with daytime concentrations significantly higher than nighttime concentrations ( $p < 0.01$ ). The Pearson's correlation coefficient for home-site indoor and outdoor concentrations was highly significant ( $r = 0.56$ ,  $p < 0.01$ ). The similar diurnal patterns in both outdoor and indoor ozone concentrations and this relatively high correlation strongly suggest that ozone in homes originates primarily from outdoor sources.

### Characterization of Indoor/Outdoor Ratios

The differences between outdoor and indoor concentrations, especially inside homes, may significantly affect exposures (20,21). Characterization of the I/O ratio is therefore important. The I/O ratio is a crucial parameter for determining penetration rate of ozone and for estimating indoor ozone concentrations when indoor measurements are otherwise unavailable.

We calculated I/O ratios for each home for 12-hr daytime periods. (Nighttime I/O ratios were not determined because the indoor concentrations were well below the LOD.) Ratios were determined using the measured 12-hr indoor concentrations ( $C_i$ ) divided by the measured 12-hr outdoor home concentrations. When measured 12-hr outdoor concentrations were not available, we estimated them using the expression:

$$C_o = C_{o24} \left( \frac{C_c}{C_{c24}} \right) \quad (1)$$

where  $C_o$  is the estimated 12-hr outdoor concentration at the home site,  $C_{o24}$  is the measured 24-hr average outdoor concentrations at the home sites, and  $C_c$  and  $C_{c24}$  are the measured 12-hr and 24-hr average outdoor concentrations at the SAM site, respectively. The I/O ratios for homes ranged from 0.07 to 1.16, with grand a mean of  $0.45 \pm 0.23$  (Table 4). The mean I/O ratios for homes, however, differed significantly by home according to ANOVA results ( $F = 3.76$ ,  $p < 0.01$ ). When excluding observations from air-conditioned homes, significant differences in mean I/O ratios among homes were still found ( $F = 3.72$ ,  $p < 0.01$ ).

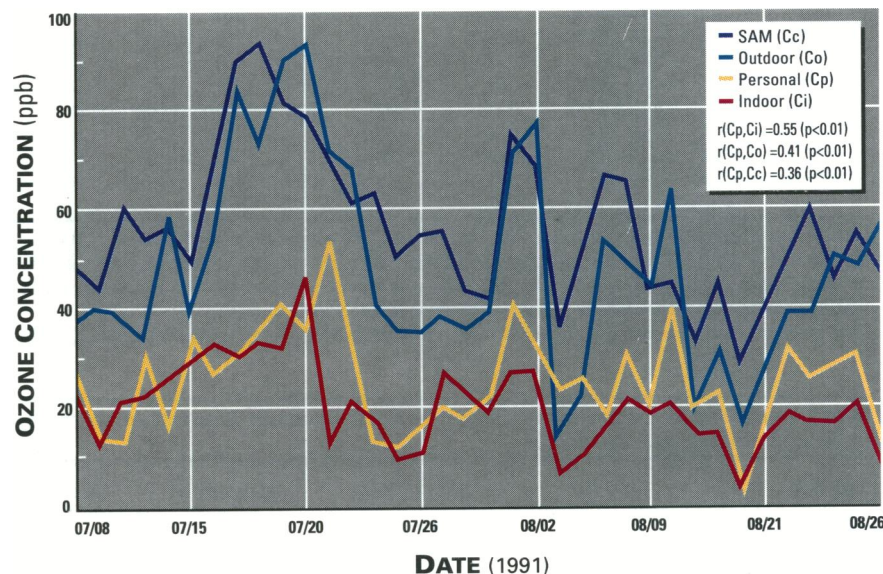
We examined information on home ventilation conditions to help understand factors that may affect I/O ratios. Three

**Table 3.** Mean distance of homes from the SAM site, mean altitude, and mean ratio of home outdoor to SAM site ozone concentration in different residential regions

Region	Distance (km)	Altitude (m)	N	Mean $\pm$ SD	Minimum	Maximum
1*	6	361 $\pm$ 9	14	0.66 $\pm$ 0.13	0.47	0.85
2	3	388 $\pm$ 18	20	0.79 $\pm$ 0.24	0.33	1.30
3	6	348 $\pm$ 6	17	0.91 $\pm$ 0.22	0.63	1.34
4*	8	352 $\pm$ 18	28	0.73 $\pm$ 0.24	0.39	1.44
5	9	237 $\pm$ 33	8	0.77 $\pm$ 0.33	0.35	1.30
6	13	365 $\pm$ 20	14	1.03 $\pm$ 0.21	0.66	1.40

Region 1 is most populated and region 6 is least populated.

\*Significant concentration differences from region 6.



**Figure 3.** Daytime ozone measurements from the stationary ambient monitoring site continuous analyzer, home outdoor, home indoor and personal passive samplers. Mean concentrations from different homes or participants for each day were plotted.

factors were considered: percentage of open windows in the house, amount of time the windows were open, and air conditioner use. During daytime periods, 98% of the windows in the homes were open for a mean period of 11.5 hr. Three households used air conditioners for an average period of 0.3 hr. Weak correlations were found for I/O ratios with the amount of time the windows were open ( $r = 0.19$ ,  $p = 0.10$ ). Use of air conditioning was not correlated with I/O ratios ( $r = 0.07$ ,  $p = 0.57$ ), which may be due to the small sample size for air-conditioned homes. Results suggest that I/O ratios may increase with greater air flow through the home. In addition, dissimilar housing materials, such as painted walls, furniture, drapes, and books, may affect ozone decay and as a result affect I/O ratios (28–31).

### Comparison of Ozone Measurements

Daytime personal exposures (or  $C_p$ ) were correlated with daytime concentrations measured inside ( $r = 0.55$ ,  $p < 0.01$ ) and outside ( $r = 0.41$ ,  $p < 0.01$ ) the home and at

the SAM site ( $r = 0.36$ ,  $p < 0.01$ ; Fig. 3). The  $C_p$  values, however, were significantly higher than the corresponding indoor concentrations ( $p < 0.05$ ) and significantly lower than both home outdoor ( $p < 0.01$ ) and SAM site outdoor concentrations ( $p < 0.01$ ) using paired  $t$ -tests. The mean ratio of personal measurements to home indoor measurements was  $1.69 \pm 3.03$ , indicating that home indoor measurements underestimated personal exposures by 41% on average [i.e.,  $100\%(1-1.69)/1.69$ ]. The mean ratios of personal measurements to the estimated daytime home outdoor values and the SAM site measurements were  $0.59 \pm 0.52$  and  $0.44 \pm 0.29$ , respectively. The ratios imply that outdoor home measurements would overestimate personal exposures by 69% on average. This overestimate would be greater (127% on average) if SAM site measurements were used to approximate personal exposures. Ratios are summarized in Table 5.

### Personal Exposure Models

We developed two types of daytime personal exposure models. Both models used

**Table 4.** Ratio of indoor to outdoor ozone concentrations for homes

Home ID	N	Mean ± SD	Minimum	Maximum	A/C	%WO	Dur	Type	Fan
1	3	0.44 ± 0.16	0.32	0.62	0	100	12	S	0
2	3	0.51 ± 0.20	0.28	0.65	0.7	100	11	S	1
4	4	0.54 ± 0.06	0.48	0.61	0	100	12	S	1
5	4	0.31 ± 0.15	0.08	0.43	3.5	90	7	A	0
6*	4	0.72 ± 0.30	0.55	1.16	0	100	12	S	0
7*	4	0.23 ± 0.15	0.10	0.44	0	100	12	S	0
8	2	0.53 ± 0.05	0.50	0.57	0	100	11	S	1
9	2	0.21 ± 0.06	0.16	0.25	0	100	12	S	1
10	5	0.53 ± 0.21	0.33	0.83	0	94	13	S	1
11	4	0.38 ± 0.30	0.13	0.79	0	100	12	S	0
12*	5	0.71 ± 0.25	0.46	1.03	0	100	12	S	1
13*	5	0.20 ± 0.09	0.07	0.28	0	100	11	S	1
14*	5	0.74 ± 0.13	0.52	0.85	0	100	12	S	2
15*	5	0.26 ± 0.10	0.15	0.40	0	100	12	A	0
18	1	0.48 ± 0	0.48	0.48	0	100	11	S	1
19	4	0.48 ± 0.22	0.27	0.77	0	85	12	S	1
20*	5	0.30 ± 0.10	0.17	0.42	2	100	12	S	0
21	4	0.53 ± 0.13	0.35	0.66	0	94	12	S	1
22	2	0.24 ± 0.17	0.12	0.36	0	100	8	S	0
23	1	0.52 ± 0	0.52	0.52	0	100	10	S	0

Samples from homes 3, 16, and 17 were void. A/C denotes use of air conditioning in hours. %WO and Dur are percent windows open and the amount of time windows were open (in hours). Type denotes the type of house: S denotes single house and A represents apartment complex. The column Fan lists number of fans used in the sampling room. All are average values for the sampling week.

\*The mean I/O ratios for homes 6, 12, and 14 were significantly different from those for homes 7, 13, 15, and 20.

**Table 5.** Ratio of daytime personal concentration to daytime home indoor, home outdoor, and the stationary ambient monitoring site concentration

Variable	N	Mean ± SD	Minimum	Maximum
$C_p/C_i$	77	1.69 ± 3.03	0.02	26.10
$C_p/C_o$	72	0.59 ± 0.52	0.01	3.44
$C_p/C_c$	74	0.44 ± 0.29	0.01	1.35

$C_p$ , 12-hr average personal concentration;  $C_i$ , 12-hr average home indoor concentration;  $C_o$ , Estimated/measured 12-hr average home outdoor concentration;  $C_c$ , 12-hr average concentration at the SAM site.

the measured daytime home indoor, personal, measured/estimated home outdoor concentrations, and the daytime time-activity information to estimate exposures. The first type of models were constructed using stepwise linear regression techniques to determine the relative influences of indoor and outdoor concentrations and time-activity patterns on personal exposures. The measured or estimated daytime home outdoor concentrations ( $C_o$ ), the measured daytime home indoor concentrations ( $C_i$ ), the fraction of time spent outdoors ( $F_o$ ) within the daytime sampling period, and the interaction terms,  $C_i(1-F_o)$  and  $C_o(F_o)$ , were included as independent variables in the model. Note that the fraction of time spent indoors ( $F_i$ ) was indirectly included in the regression analysis because it is inversely correlated with the fraction of time spent outdoors ( $F_i = 1-F_o$ ). Concentrations of all indoor microenvironments were assumed to equal those measured inside the homes.

The stepwise variable selection technique suggests that indoor ozone concentrations ( $C_i$ ) were the most significant predictors of personal exposures (Table 6, model 1). This is not surprising, given the strong association between these two variables ( $r = 0.55$ ). The other important predictor variable added in the model was the interaction term  $C_o \times F_o$  (Table 6, model 2), suggesting that outdoor ozone concentrations were predictive when weighted by the fraction of time spent outdoors. Model 2 explained 37% of the variability in personal exposures and had a slightly smaller relative mean standard error than model 1. We tried different variable selection techniques for this analysis, including forward stepwise and backward elimination procedures based on  $F$  statistics for a variable's contribution to the model. Procedures based on maximizing the adjusted  $R^2$  statistic were also performed. Each of these procedures leads to the same final model 2.

Because the results from the above statistical models do not have an intuitive

interpretation, we constructed a second type of model based on the microenvironmental exposure concept (32–34). A simple prediction of daytime personal exposures ( $C_e$ ) is the time-weighted average of the outdoor and indoor exposures:

$$C_e = [C_i(1-F_o)] + [C_o(F_o)] \quad (2)$$

The multiple regression model incorporating an intercept term is summarized as model 3 in Table 6. This model has a similar fit to that of model 2 ( $R^2 = 0.35$ , root mean squared error = 13.68) and has a nonsignificant intercept of  $4.67 \pm 3.70$  ( $p = 0.21$ ).

Because children generally spend time outdoors when ambient ozone concentrations are highest, the time of day a child is outdoors may be an important determinant of personal exposures. To incorporate this factor into the time-weighted model, we divided concentration and activity data into 1-hr intervals. We estimated hourly outdoor and indoor daytime concentrations using continuous measurements from the SAM site. Hourly outdoor concentrations ( $C_{o,k}$ ) were estimated for each home using the expression:

$$C_{o,k} = \frac{C_{o24}}{C_{c24}}(C_{c,k}) \quad (3)$$

where  $C_{o24}$  is the 24-hr outdoor ozone concentration measured at home sites,  $C_{c24}$  is the 24-hr outdoor ozone concentration measured at the SAM site, and  $C_{c,k}$  is the daytime 1-hr outdoor ozone concentration measured at the SAM site at hour  $k$ . Hourly indoor concentrations ( $C_{i,k}$ ) were estimated in a similar way:

$$C_{i,k} = \left(\frac{C_i}{C_o}\right) \left(\frac{C_{o24}}{C_{c24}}\right) \times (C_{c,k}) \quad (4)$$

where  $C_i/C_o$  is the indoor/outdoor ratio. Then, the daytime hourly microenvironmental model is as follows:

$$C_e = \sum_{k=1}^{12} \frac{[C_{i,k}(1-F_{o,k})] + [(C_{o,k})(F_{o,k})]}{12} \quad (5)$$

where  $F_{o,k}$  is the fraction of time spent outdoors in the  $k$ th hour.

When personal exposures ( $C_e$ ) estimated by the model were regressed on measured personal exposures ( $C_p$ ), the hourly microenvironmental model (model 4 in Table 6; Fig. 4) explained a slightly higher percentage of the variability in measured personal exposures ( $R^2 = 0.40$ ) and had a smaller root mean squared error than the 12-hr simple microenvironmental model (model 3).

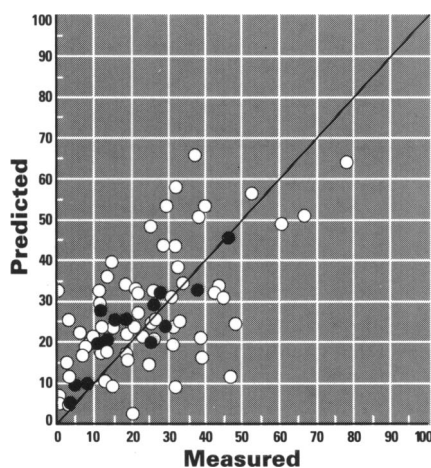
Further improvements in the predictive power of the hourly microenvironmental



ozone model may be achieved by accounting for the contribution of diverse outdoor and indoor microenvironments to personal ozone exposures. We anticipated that the regression model would have the most predictive power for those days on which individuals spent most of the time indoors and outdoors near the home for which corresponding exposure measurements were available. Support for this hypothesis is evidenced by the fact that model 5 predicted exposures more accurately for participants who spent at least 95% of the day in or near their homes than for those who did not (Fig. 4). When we fitted the analogue of model 5 (Table 6) only to the 14 observations from participants who stayed at or near their home for at least 95% of the monitoring period, 76% of the variability in personal ozone exposures was explained.

## Discussion and Conclusions

Results from this pilot study indicate that the traditional method of using fixed-site measurements to represent individual exposures may not be appropriate. Our results showed a significant spatial variation in outdoor ozone concentrations for a small college town, with densely populated regions having lower ozone concentrations than rural regions. The spatial variation may be due to differences in density of houses and/or population, traffic intensity, and availability of NO sources. Ignoring the spatial variation and using the fixed-site measurements alone to estimate personal exposures can result in an error as high as 127%. Had the SAM site been located in one of the residential areas of town, the error in personal exposure estimates may have been lower.



**Figure 4.** Predicted personal exposures using estimated hourly home indoor and home outdoor concentrations versus measured personal exposures. Shaded dots represent observations from children who spent at least 95% of the day in or near their homes. Note that the graph is overlaid by a 45° line.

**Table 6.** Regression models for predicting personal ozone exposures

Model	Covariate	Coefficient	$p$	RMSE	$R^2$	$N$
Stepwise regression						
1	Intercept	$8.74 \pm 3.36$	0.01	14.09	0.30	68
	$C_i$	$0.76 \pm 0.14$	<0.01			
2	Intercept	$5.27 \pm 3.44$	0.13	13.44	0.37	68
	$C_i$	$0.67 \pm 0.14$	<0.01			
	$C_o \times F_o$	$0.39 \pm 0.14$	<0.01			
Microenvironmental						
3	Intercept	$4.67 \pm 3.70$	0.21	13.68	0.35	68
	$C_i \times (1 - F_o)$	$0.77 \pm 0.17$	<0.01			
	$C_o \times F_o$	$0.62 \pm 0.14$	<0.01			
4	Intercept	$5.16 \pm 2.99$	0.09	12.48	0.40	79
	$C_e$	$0.70 \pm 0.10$	<0.01			
5	Intercept	$-4.22 \pm 4.27$	0.34	6.41	0.76	14
	$C_e$	$1.05 \pm 0.17$	<0.01			

RMSE, root mean squared error. *C*<sub>i</sub>, 12-hr average personal concentration; *C*<sub>o</sub>, 12-hr average home indoor concentration; *C*<sub>o</sub>, estimated/measured 12-hr average home outdoor concentration; *C*<sub>e</sub>, 12-hr average concentration at the standard ambient monitoring site.

Indoor/outdoor (I/O) ratios varied by home, with typical mean ratios ranging from 0.20 to 0.74. Results from simple regression and correlation analyses suggest that the I/O ratio differences may be due partly to house ventilation conditions and dissimilar housing materials. Other studies also have shown that the I/O ratios of other indoor microenvironments vary widely. For example, Thompson et al. (17) showed that in hospitals, the mean I/O ratios for total oxidants ranged between 0.5 and 0.67. In office buildings, mean I/O ratios ranged from 0.3 to 0.8 (17,20,35–37), while the mean I/O ratios were approximately 0.3 and 0.6 for air-conditioned and non-air-conditioned school classrooms, respectively. For a large shopping mall, where outside air was minimal, the mean I/O ratio was approximately zero (17).

As a result of the outdoor spatial and indoor concentration variations, the ability to predict personal exposures from outdoor and indoor concentrations was poor (*r*<sup>2</sup> = 0.35), even when time-weighted concentrations were used (*r*<sup>2</sup> = 0.40). The inability of the simple microenvironmental model to estimate personal exposures may have resulted from the consideration of only two microenvironments, indoor home and outdoor home, by the model. However, when activities were limited to locations in or near the home, the accuracy of the simple microenvironmental model improved substantially (*r*<sup>2</sup> = 0.76). It is evident that contribu-

tions from diverse indoor and outdoor microenvironments must be considered to estimate personal ozone exposures accurately.

To improve our ability to model personal ozone exposures, future studies should characterize indoor and outdoor concentrations in a variety of indoor and outdoor microenvironments within the same community. This effort should examine factors that affect indoor and outdoor ozone concentrations. For indoor concentrations, these factors may include air exchange rates, housing materials, gas stove use, home volumes, home interior surface type. For outdoor concentrations, the effects of NO sources and/or traffic density, house density, and population density should be investigated. In this regard, we have continued investigating factors affecting variations in indoor and outdoor ozone concentrations. In the Canadian Research on Exposure Assessment and Modeling Project (38), we measured outdoor ozone concentrations at different locations in Toronto, Canada, and collected indoor ozone samples in a variety of indoor environments, such as schools, office buildings, and retail stores. The results from this study will be presented in forthcoming papers (Liu et al., in preparation).

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